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Invention: Glass Compositions				
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Filing Date	July 25, 2002
First Named Inventor	Brian Algar
Art Unit	1614
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Attorney Docket Number	7678.576a.1

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**2. Patent application number***(The Patent Office)***9919283.3**

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If the applicant is a corporate body, give the country/state of its incorporation

Brian Algar  
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United Kingdom

77206 83001

**4. Title of the invention**

A glass composition.

**5. Name of your agent *(if you have one)***"Address for service" in the United Kingdom to which all correspondence should be sent *(including the postcode)*

ROYSTONS,

Tower Building,  
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Title: A glass composition.

### DESCRIPTION

The present invention relates to a glass composition, particularly but not exclusively for the improved treatment and/or prevention of dental caries.

Dental caries consists of demineralization of a tooth caused by bacteria. In the early stages of caries a white spot develops on the tooth and if the disease is not halted and reversed, the enamel surface breaks down to form a lesion. This can then lead to decay and eventually, a fractured tooth. It is well known that development of dental caries may be reduced by means of various factors, such as diet and oral hygiene measures, anti microbial treatments and the provision of fluoride to the teeth.

Current methods for administering fluoride include the fluoridation of drinking water, the ingestion of fluoride tablets, the incorporation of fluoride into mouth washes, dentifrices and foods, the topical application of fluoride solutions, gels and varnishes and recently, the incorporation of fluoride in dental materials and special devices. These have a variable effect on caries which is unpredictable on an individual basis and is dependent on patient compliance in following the prescribed regimen.

Evidence supports the concept of frequent applications of relatively low concentrations of fluoride ions for the elimination of caries. A sustained and controlled release delivery system could help to achieve this goal. At least three general approaches have been reported for the application of sustained and controlled slow releasing systems, being a sustained release ingested tablet or capsule (Masuhara et al 1985), incorporation of fluoride in dental cements (McClean & Wilson) and an intra-oral device attached to the teeth (Minth et al 1983). However, none of these devices has

proved to be suitable for use. They have either been susceptible to damage, an irritant to the mucosa or non acceptable to the patient.

Glass compositions for attaching to a tooth that release fluoride ions in the mouth to supplement dietary intake of fluoride have proved useful, where normal intake levels of fluoride are insufficient to give maximum reduction in the incidence of caries lesions in teeth.

A glass from which fluoride can be slowly leached was patented by Davidson (US Pat. No. 4,920,082). The glasses described therein consist of silicon dioxide, barium oxides, aluminium oxide and fluoride in specified ranges. However, the maximum fluoride which can be retained in this system is 7% by weight and batch melting temperatures in the range of 1300-1400°C are generally required. WO88/05652 also describes the preparation of novel dental composites that are claimed to release fluoride, incorporating fluorosilicate glass fillers, the glass consisting essentially in weight percent of 15-50%  $\text{Al}_2\text{O}_3$ , 0-50%  $\text{CaO}$ , 10-65%  $\text{SiO}_2$  and 0-14% F. Again, silicate glass is known to melt at high temperatures which is unfavourable.

The use of phosphate as a glass former has been known for many years. However, the disadvantage of these glasses is that they are easily attacked by water. This property has been exploited for the development of soluble glasses for use in animal health releasing copper, cobalt and selenium to the ruminant animal over 6 to 12 months as the glass dissolves (GB Pat. No. 2116424). A more slowly dissolving glass has been used to provide copper ions in an anti-fouling paint for use on ships. This glass was formulated to dissolve over 5 years (EP App. No. 94906287.1).

Hence, the glass compositions of the prior art have not proved entirely satisfactory for supplementing the dietary intake of fluoride. The low retention of the fluoride means that the release of fluoride is not maintained over a sufficiently long period of time. The low retention also requires a relatively large piece of glass to be fixed to the tooth of the patient to provide sufficient levels of fluoride release into the mouth. This is obtrusive and reduces the appeal of the device to a patient.

It is an aim of the present invention to provide a glass composition for the improved treatment and/or prevention of dental caries that aims to overcome the above mentioned drawbacks.

Accordingly, a first aspect of the present invention provides a glass composition having the general empirical formula given below, expressed in weight percent of the element:

P : 16-24

F : 5-30

O : 20-40

and at least one of Na, K, Li or Al in an amount up to a total of 34 wt.%.

Preferably, fluoride and/or oxides of glass modifiers, such as Al, Ca, and Mg, are included in the composition. The fluoride ions are preferably included as compounds such as  $\text{AlF}_3$ ,  $\text{NaHF}_2$ ,  $\text{NaF}$ ,  $\text{CaF}_2$ ,  $\text{MgF}_2$  or  $\text{KF}$ .

Ca and Mg and/or other glass modifiers are preferably included in the composition in an amount 0-10 wt.%, more preferably less than 5 wt.%

The glass compositions of the present invention preferably provide a fluoride retention of at least 50% at a melting temperature of  $650^\circ\text{C}$  over 45 minutes, preferably



at least 60%. Preferably, the glass composition has a low solubility rate thereby allowing fluoride release from the composition for a period of 12-36 months. The glass compositions of the present invention may be attached to a tooth, for example being attached to a rear molar using standard dental cement or as a powder for adding to dental restorative materials, such as dental amalgams, thereby providing means to supplement fluoride release into saliva to assist in the prevention or reduction of dental caries.

More preferably, the composition includes a combined weight percent of at least 18% of sodium and potassium, more preferably 20-26 wt.%. Al is preferably included in an amount of at least 4 wt.%, more preferably 4-7 wt.%.

The composition preferably has at least 25 wt.% oxygen, more preferably 25-35 wt.%, has at least 16 wt.% phosphorous, more preferably 18-22 wt. %, and has at least 12 wt.% F, more preferably 15-25 wt.%.

The phosphorous may be included in the composition as an oxide, such as  $P_2O_5$ . The alkali metal compounds may be included as, for example, their oxides or fluorides.

The present invention will now be further illustrated by means of the following Examples in which Example 1 investigates the percentage fluoride retention for 25 samples of glass compositions according to the present invention and Example 2 investigates the percentage of fluoride in saliva following attachment of a glass composition of the present invention to a patient's tooth, and with reference to the accompanying drawing in which the percentage fluoride retention of the 25 samples investigated in Example 1 is illustrated.

### **Example 1**

Table I of the accompanying drawing illustrates the composition parameters of 25 glass compositions according to the present invention, labelled 1-25 respectively. The percentage fluoride retained by each composition was calculated from determining the theoretical fluoride percentage of each sample and comparing this to the analysed level of fluoride found after the glass had been melted. The compositions were recorded as weight percentages of the elements. This was done to ensure the most accurate recording of percentage of fluoride retained. The method often used that involves recording the elements present as oxides and then recording the fluorine as a separate element is incorrect as the fluorine ions are taking the place of oxygen in the glass matrix. Assigning the fluoride ion to any particular element is also incorrect, as the actual location of the fluoride ion is unknown. The method used herein was therefore preferred and conversion to other older systems is easily achieved for comparison purposes.

The percentage fluoride retained by the samples is given in Table I of the accompanying drawing.

### **Example 2**

The percentage of fluoride released into saliva following attachment of a glass composition of the present invention to a tooth was investigated.

The batch components using compositions according to the present invention were thoroughly mixed to ensure a homogeneous melt and loaded into platinum crucibles. The crucibles were then placed in an electric melting furnace at temperatures from 600-650°C, for times of up to 60 minutes to achieve good melting. The crucibles

were removed from the furnace and the glass cast onto a moulding plate containing a number of circular holes of 4mm in diameter and 6mm in depth. The glass was rolled to force it into these cavities and when solidified it was removed from the plate and transferred to an annealing oven to slowly cool and remove any residual stress.

Before use, the devices were smoothed of any rough edges with a sharp diamond burr. Three tests were carried out with human subjects and the results are reported in Tables II – IV below. The device was attached to the buccal aspect of the first maxillary permanent molar because of its nearness to the opening of the parotid gland. It was felt that the salivary flow would help to distribute the fluoride to other parts of the mouth. The tooth was cleaned using a fluoride-free prophylaxis paste. After cleaning, the tooth was washed, dried and the buccal surface etched for one minute with the etch available in the composite kit. (Prisma Fil-Predosed High Density Composite, The L.d. Chalk Company, Division of Dentsply International Inc., Milford, Delaware 19963, U.S.A.). The glass was etched for twenty seconds. Both were washed, dried and a thin layer of a light cured bond from the composite kit brushed on the tooth and the glass. The glass, held by tweezers, was adapted to the tooth and cured by visible light. While temporarily held in place, a layer of light cured composite was adapted around the glass using a plastic instrument. This composite helps to retain the glass, blocks out any under cuts and makes the whole device smooth to the tongue. Once cured, the glass and composite were further smoothed with a white stone burr and a layer of fissure sealant placed on the composite and cured to give a smoother surface. Control of moisture was very important throughout this procedure. Care was taken not to cover the exposed releasing surface of the glass with any composite, bond or fissure sealant.

In these studies, efforts were made to ensure that the fluoride in saliva could be accurately and repeatedly determined. The method employed was that of Taves (Separation of fluoride by rapid diffusion using hexamethyldisiloxane, *Talanta* 15, 969-974, 1968), in which fluoride was diffused from the samples using hydrochloric acid saturated with hexamethyldisiloxane  $(\text{CH}_3)_3\text{SiOSi}(\text{CH}_3)_3$  (HMDS). Fluoride was collected in sodium hydroxide before its determination by a fluoride ion electrode.

For each sample of saliva, between 1.0 and 2.0g (depending on the sample size) was weighed into a 60x15mm polystyrene petri dish (Falcon Plastics Cat No. 1007, Fahrenheit Lab. Supplies, Leeds). This avoided the difficulty of trying to accurately pipette this viscous material. Distilled water was added to make the final volume of 3.0ml. Polystyrene tube caps (Falcon Plastics Cat No. 2051, Fahrenheit Lab. Supplies, Leeds.), with the rims reduced by a half, were placed in the centre of each dish.

0.1ml of 1.65M NaOH containing P-nitrophenol and phenolphthalein as an indicator was added to the centre of each well to ensure that the trap remained alkaline and therefore was able to trap the fluoride. An alkaline trap remained pale yellow in colour while an acidified trap turned pink. The lids were then sealed on the petri dishes using petroleum jelly around the rims. Finally 1.0ml of 6.0M HCl with HMDS was added to each dish via a small hole previously made in the lid and the hole sealed immediately with petroleum jelly and a square of sealing tissue.

The samples were placed on a rotary shaker at 200 rotations /minute and left to diffuse overnight which was usually 16 hours. The following morning the lids were prized off and a note made of any of the dishes which had not formed a vacuum since loss of the vacuum had been shown in preliminary tests to be an indicator that fluoride had been lost

from the system. Each of the caps were removed and placed in an oven at 100°C until the NaOH had become crystalline.

After drying the NaOH the caps were placed on their test tubes and shaken with 0.34M acetic acid to dissolve the crystals and bring the pH to 5.2 for its determination by a combination fluoride ion electrode. Fluoride standards of 0.05, 0.1, 0.5, 1.0 and 5.0 ug/ml F were prepared in identically buffered solutions to the sample solutions and were used to construct a standard curve. The fluoride concentrations in the unknown diffused samples were measured from this curve using an Orion combination fluoride ion electrode and Orion 920A Ionanalyser (Orion Research Inc., Cambridge, MA.). From the concentrations of the diffused solutions, the concentrations of fluoride in the original sample were calculated.

In all analyses, known fluoride standards and blanks were also diffused to determine the percentage diffusion that was occurring. The level of diffusion varied between 96.0 and 108% where a vacuum had been maintained. When known samples of fluoride solutions were diffused, the standard deviation between the recoveries was usually less than 1.0%.

Fluoride blanks were also run in this system. These included HCl-HMDS + water or NaOH alone. Fluoride was not measurable in the water or the HCl after it had been saturated with the HMDS. It appeared that the NaOH was the main contributor to the small blank of 0.002 ug/ml F.

A check was made to determine if the fluoride concentration changed if the analyses were not done on the day of collection since it was possible that there would be too many samples to handle on some of the days when a study of salivary fluoride was

being carried out and/or because laboratory facilities were not available every day.

Therefore, duplicate analyses were carried out on samples which had been kept up to 7 hours at room temperature and on samples which had been stored up to 10 days at  $-12^{\circ}\text{C}$ .

The former was to check if study subjects could take part in collecting saliva while going about their normal work and bring the samples for analysis at the end of the collection period. The latter was to allow for the collection and storage of saliva samples when laboratory facilities were not available or for when the numbers of samples had built up beyond those which could be readily handled. No differences were found in the fluoride levels analysed immediately, after up to 7 hours or after 10 days at  $-12^{\circ}\text{C}$ .

The recovery of fluoride and reproducibility of the method were checked by using known standards and by carrying out repeat analyses on the same samples. The results of these preliminary analyses are shown in Table II below.

**Table II**

**Reproducibility and Recovery of Fluoride After Diffusion of Samples in HMDS-HCl**

Sample		No of Tests	Known $\text{F}^1$	Determined F		Recovery
No 2	(ml solution)		$\text{F}^-$	$\text{F}^-$	$\text{SD}^3$	
1	1	4	0.05	0.045	$\pm 0.002$	90
2	1.0	4	0.1	0.092	$\pm 0.010$	92
3	0.5	6	0.5	0.475	$\pm 0.030$	95
4	0.5	3	1.0	0.940	$\pm 0.010$	94
5	0.2	2	5.0	4.900	$\pm 0.008$	98
6	Dist $\text{H}_2\text{O}$	10	NIL	$<0.005$	$\pm 0.000$	
7	Whole resting saliva	6	Unknown	0.010	$\pm 0.003$	
8	Whole resting saliva	10	Unknown	0.015	$\pm 0.005$	

<sup>1</sup> F =	Fluoride concentration in ug/ml
<sup>2</sup> Samples	1,4 and 7 were from one standard solution or one saliva source and analysed at the same time. 2, 5, and 6 were from known solutions or distilled H <sub>2</sub> O prepared and analysed at different times. 3 and 8 were from one solution or one saliva source analysed at different times.
<sup>3</sup> SD =	Standard deviation.

The results show that the recovery of fluoride by the test system was better than 90%. There was therefore an error of only 10%, which was thought to be very good, and the system was acceptable.

Table III below shows the fluoride concentration in saliva of a subject fitted with a device made from glass composition 16 in Table I of the accompanying drawing. A marked high increase in fluoride level in saliva was observed within the first three days. This level then settled down to a steady 0.025 - 0.035 ug/ml., giving on average a three-fold increase from the 0.09 - 0.12 ug/ml base line. The effect of the device was still clearly seen even after a year and a half in place in the subject.

**Table III****Fluoride Concentration in  $\mu\text{g/ml}$ . of saliva for subject fitted with glass 16**

		Fluoride Concentration $\mu\text{g/ml}$
Base line	Day 1	0.012
	Day 2	0.009
Treatment	Day 1	0.063
	Day 2	0.039
	Day 3	0.069
	Day 4	0.031
	Day 5	0.028
	Day 6	0.037
	Week 1	0.036
	Week 2	0.025
	Week 3	0.024
	Month 1	0.030
	Month 2	0.030
	Month 3	0.036
	Month 4	0.038
	Month 5	0.034
	Month 6	0.033
	Year 1	0.03
	Year 1.5	0.05

Table IV below shows the averaged fluoride concentration in saliva of two subjects fitted each with a device made from composition 14 in Table I of the accompanying drawing. A marked high increase in fluoride level in saliva was again observed at the start but this time only for the first two days. This level then settled down to a steady 0.02 - 0.025  $\mu\text{g/ml}$ , giving on average a two and a half fold increase from the 0.009  $\mu\text{g/ml}$  base line. The device was still effective up to the four month point when it was removed.



**Table IV**

**Averaged Fluoride Concentration in  $\mu\text{g/ml}$ . of saliva for two subjects  
fitted with glass 14**

		Fluoride Concentration $\mu\text{g/ml}$ .
Base Line	Day 1	0.009
	2	0.009
Treatment	Day 1	0.056
	2	0.053
	3	0.023
	4	0.024
	5	0.024
	6	0.035
	Week 1	0.025
	2	0.022
	3	0.023
	Month 1	0.025
	2	0.017
	3	0.022
	4	0.022

Table V below shows the averaged fluoride concentration in saliva of four subjects fitted each with two devices made from glass composition 14 in Table I of the accompanying drawing. Again the level of fluoride was high for the first three but then settled down to around the 0.035  $\mu\text{g/ml}$ ., a level which was a three and a half fold increase over the base line. There was also an unusually high level of fluoride recorded at weeks 1 and 2.

**Table V****Averaged Fluoride Concentration in  $\mu\text{g/ml}$  of saliva for four subjects****each filled with two devices of glass 14**

		Fluoride Concentration $\mu\text{g/ml}$
Base Line	Day 1	0.01
	Day 2	0.01
Treatment	Day 1	0.043
	2	0.053
	3	0.045
	4	0.034
	5	0.041
	6	0.032
	Week 1	0.067
	2	0.054
	3	0.033
	Month 1	0.036
	2	0.033
	3	0.035
	4	0.033
	5	0.035
	6	0.032

The results clearly show that high levels of fluoride can be retained by the glass compositions of the present invention, up to 15% in the compositions included in Table I, with acceptable levels of fluoride retention (up to 67% in the composition of Table I). Additionally, melting temperatures can be kept low, generally at  $650^{\circ}\text{C}$  or less. The glass compositions also allow fluoride to be released from the glasses for up to at least  $1\frac{1}{2}$  years, as illustrated in Table III. Furthermore, the level of fluoride released from the composition can be adjusted by changes in retained fluoride, glass composition and the number of devices used, as shown in Table I and III to V. Hence, the compositions of the present invention provide a glass containing fluoride for insertion into the mouth for a slow

continuous release of fluoride ions, the glass being fitted either by attachment of a formal article of the glass to a tooth, being held in place by a dental plate or by incorporation of glass in a powder form as a dental restorative.

**Table I**

**Glass Compositions and Analyses**

Code	Na	P	Al	F	O	K	Ca	Mg	Na+ K	F Analysed	Solubility	Melt Temp	Melt Time	Fluoride Retention
1	21.23	20.67	6.76	19.48	31.86	0	0		21.23	10.4	149	650C	45MIN	53.39
2	22.47	20.37	6.22	19.54	31.39	0	0		22.47	10.97	503	650C	45MIN	56.14
3	23.68	20.08	5.69	19.6	30.94	0	0		23.68	11.81	2920	650C	45MIN	60.26
4	21.54	21.18	6.26	18.37	32.64	0	0		21.54	10.94	288	650C	45MIN	59.55
5	21.16	20.81	5.27	18.94	32.07	0	0	1.75	21.16	11.45	215	650C	45MIN	60.45
6	21.23	20.67	6.76	19.48	31.86	0	0		21.23	11.42	149	650C	45MIN	58.62
7	21.13	20.87	4.55	18.67	32.17	0	0	2.59	21.13	11.78	305	650C	45MIN	63.10
8	22.88	20.27	6.04	19.56	31.25	0	0		22.88	12.01	1385	650C	45MIN	61.40
9	23.28	20.17	5.87	19.59	31.09	0	0		23.28	12.15	2513	650C	45MIN	62.02
10	23.21	19.68	6.22	20.56	30.33	0	0		23.21	11	1816	650C	45MIN	53.50
11	22.72	20.31	6.11	19.56	31.3	0	0		22.72	12.03	1962	650C	45MIN	61.50
12	22.91	19.16	6.72	21.68	29.53	0	0	0	22.91	14.11	1684	650C	45MIN	65.08
13	23.14	18.55	6.97	22.76	28.59	0	0	0	23.14	14.95	1715	650C	45MIN	65.69
14	21.9	20.13	5.27	19.94	31.02	0	0	1.75	21.9	13.22	405	650C	45MIN	66.30
15	22.63	19.44	5.26	20.94	29.97	0	0	1.75	22.63	14.06	661	650C	45MIN	67.14
16	22.13	18.76	5.26	22.3	28.92	0	0	2.62	22.13	14.79	685	650C	45MIN	66.32
17	21.83	18.24	5.75	23.42	28.12	0	0	2.64	21.83	14.86	560	650C	45MIN	63.45
18	14.92	20.67	6.76	18.04	31.86	7.75	0	0	22.67	11.15	61	650C	45MIN	61.81
19	14.29	19.79	6.25	19.71	30.5	7.68	0	1.78	21.97	12.42	100	650C	45MIN	63.01
20	22.07	21.09	6.01	18.34	32.5	0	0	0	22.07	11.2	194	650C	45MIN	61.07
21	21.85	21.68	5.76	17.29	33.42	0	0	0	21.85	10.2	574	650C	45MIN	58.99
22	18.95	19.44	5.26	20.1	29.97	4.53	0	1.75	23.48	12.6	389	650C	45MIN	62.69
23	21.23	20.67	6.76	19.48	31.86	0	0		21.23	11	130	650C	90MIN	56.47
24	21.23	20.67	6.76	19.48	31.86	0	0		21.23	10.2	79	650C	180MIN	52.36
25	18.95	19.44	5.26	20.1	29.97	4.53	0	1.75	23.48	13.41	466	600C	45MIN	66.72